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13. ABSTRACT (Maximum 200 words)

This Final technical report summarizes the most important findings of the research work on (a) lattice mismatch strain induced formation of coherent 3D islands, also dubbed self-assembled quantum dots, and (b) growth of such island quantum dots on *in-situ* growth control prepared submicron width stripe mesas. The specific accomplishments include the demonstration of (1) a re-entrant behavior of 2D to 3D island morphology evolution, (2) mass exchange between 2D and 3D features during evolution of their density, (3) quantum box nature of the 3D islands via photoluminescence excitation spectroscopy, (4) 3D island strain driven adatom migration during cap layer growth, (5) vertically self-organized growth of island quantum dots, (6) lasing from single and vertically self-organized stacks of quantum dots, and (7) selective area growth of InAs island quantum dots on sub 100nm GaAs(001) stripe mesa via control of interfacet In migration.

14. SUBJECT TERMS
Semiconductors GaAs, InAs, nanostructures, quantum dots, strained heteroepitaxy, scanning tunneling microscopy, atomic force microscopy, molecular beam epitaxy, photoluminescence, cathodoluminescence, quantum wires, quantum dot laser, self-organized growth, focused ion beam etching, growth-controlled nanostructures.

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Nanoscopic Processing and Atomistic Kinetics of Lattice Mismatched Growth on Submicron Mesas

FINAL TECHNICAL REPORT
(Apr. 1, 1993 - May 31, 1997)

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FINAL TECHNICAL REPORT (April 1, 1993 - May 31, 1997)

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I. STATEMENT OF THE PROBLEMS STUDIED

Effort under the above referenced contract was focused upon examining some *in-situ*, growth controlled approaches to synthesis of semiconductor nanostructures (quantum wires and quantum boxes) with particular emphasis on mechanisms of lattice mismatch strain accommodation and associated kinetic behavior. To this end, the following two different approaches were studied:

(1) Formation behavior of the coherent, three-dimensional (3D) islands in highly strained epitaxy and their potential use as quantum boxes.

(2) Control of density and size of such island quantum boxes via selective area growth on size-reduced patterned substrates containing appropriately oriented mesas.

Studies of the first year or so showed a great potential for the coherently strained 3D island quantum boxes on planar substrates (also dubbed self-assembled quantum dots) for optoelectronic applications. Consequently, during the next two years (mid 1994 and mid 1996) of this contract period a greater effort was placed upon this aspect and resulted in a number of new and important findings and demonstrations, including the first unambiguous demonstration of a quantum box laser. The laser comprised vertically self-organized but electronically essentially uncoupled multiply stacked quantum dots as the active region. In the following we provide a brief summary of the most important results obtained in the two categories noted above. Details may be found in the publication numbers noted for each topic in the summary. These correspond to the publication list provided in Section III.

II. SUMMARY OF THE MOST IMPORTANT RESULTS

II.1 3D Island Quantum Dots on Planar Substrates

1. *In-situ* UHV STM/AFM studies of 3D island evolution and implications for an atomistic kinetic frame work:

   Studies central to the understanding of the process and mechanism of the lattice mis-match induced coherent 3D island formation, using InAs/GaAs system as a vehicle, were carried out. The experimental findings are described in detail in publication nos. 16, 23 and 24. Briefly, we discovered that, just after the initiation of the well-formed 3D islands at ~1.57ML InAs deposition, the lateral size dispersion and average size of the islands first increases drastically (from panel (a) to (b) in Fig. 1) with about 0.05ML of additional InAs

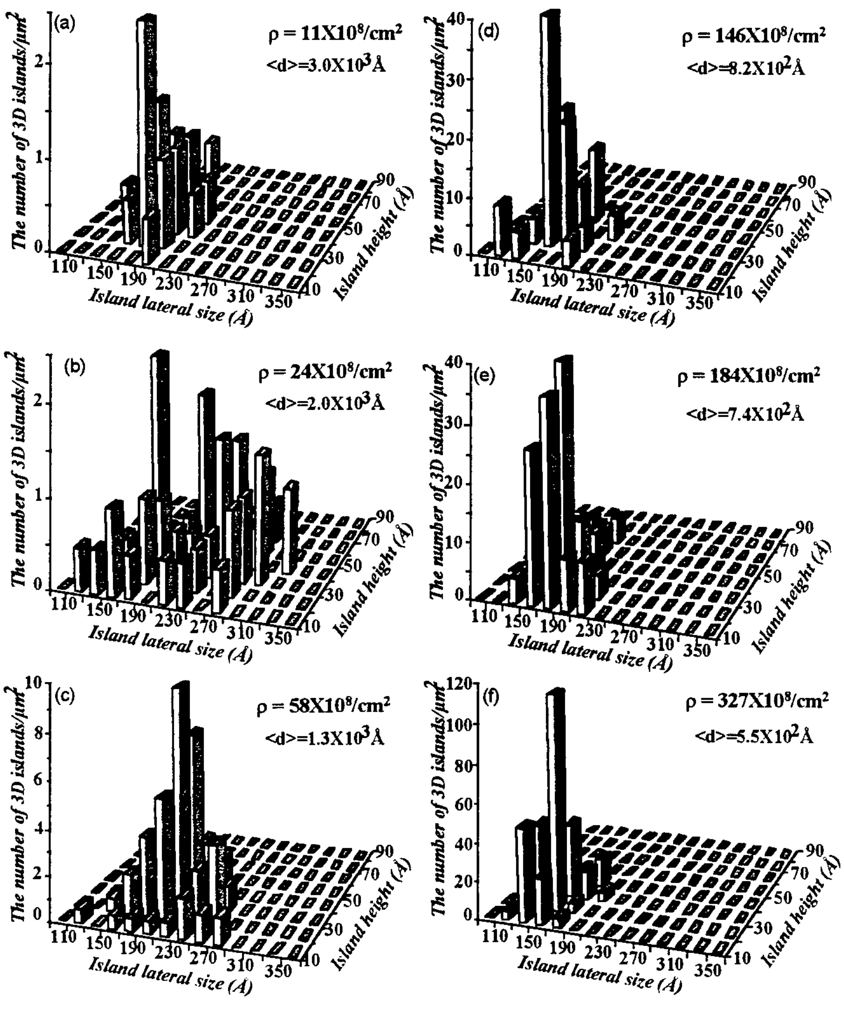

Fig. 1

deposition and then decreases and saturates (panels (c) through (f) in Fig. 1). This indicates the onset of a natural tendency for size equalization, including through loss of material from the initially formed largest islands. Moreover we discovered the presence, during the 2D-3D morphology transition, of a varying mass transfer between 2D and 3D surface features with increasing InAs deposition. The evolution of the density of different structural features is summarized in Fig.2. The quasi-3D clusters (0.6-1.2nm high, labeled as Q3D clusters in the middle panel of Fig. 2) are found to mediate the 2D-3D morphology change and to play an important role in the mass redistribution on the surface. These results provide clear evidence for the importance of the synergistic evolution of local (and hence, globally inhomogeneous) strain and surface/interface energy in determining the surface kinetic processes during the 3D island formation and evolution.

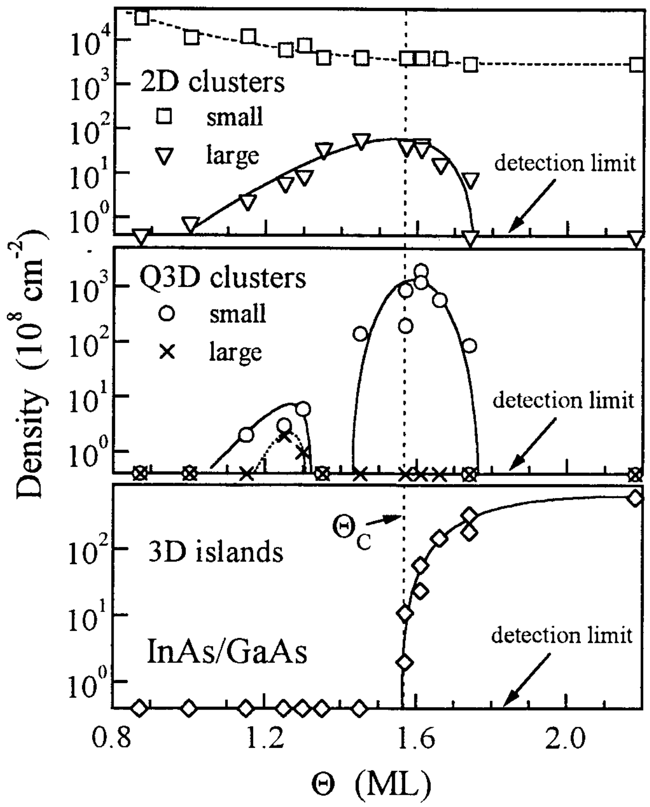

Fig. 2

2. Re-entrant behavior of 2D-3D morphology change and implications for lasing from quantum dots:

The coherent nature of the 3D islands caused explosive growth during the period of this contract in the examination of their optical behavior as quantum boxes by groups around globe and of their potential for quantum box based injection lasers by a few, including us. Reliable interpretations of the origin of optical emission and lasing demanded that careful and systematic combined atomic level structural and optical studies be carried out on comparable samples in order to understand the atomistic mechanism of strain-

induced evolution of structural features and their role in the optical response. We undertook this challenging task under the support of this contract. A remarkable discovery of a re-entrant behavior, as described below, was made and reported in a paper published in Phys. Rev. Lett. (publication no. 25).

Briefly, InAs structural features up to five monolayers high appear at ~1.25ML deposition of InAs, disappear, and reappear prior to the onset of well-developed 3D islands at 1.57ML, thus manifesting a hitherto unrecognized reentrant behavior in the formation of 3D islands (see middle panel of Fig. 2). The optical signature of this reentrant behavior is shown in Fig. 3. The narrow peak near 8500Å, which evolves with increasing InAs deposition and vanishes just beyond Θ_C at 1.57ML, is attributed to the recombination in the wetting layer (WL). The almost Gaussian peak observed at 10200Å for the 2.00 ML sample is attributed to recombination in 3D island quantum dots (QDs). A careful analysis of the PL spectra of the 1.15 ML and 1.25 ML samples reveals for the first time peaks at 9380Å and 9733Å, respectively, in addition to the WL emission. By contrast, no PL in the 9200Å to 10300Å region could be resolved for the 1.35 ML and 1.45 ML samples. And then, at 1.55 ML deposition (just below Θ_C) PL reappears in this spectral region and finally develops into 3D island PL, thus establishing a re-entrant PL behavior paralleling that of the 3D structural features seen in the STM studies. These results provide new

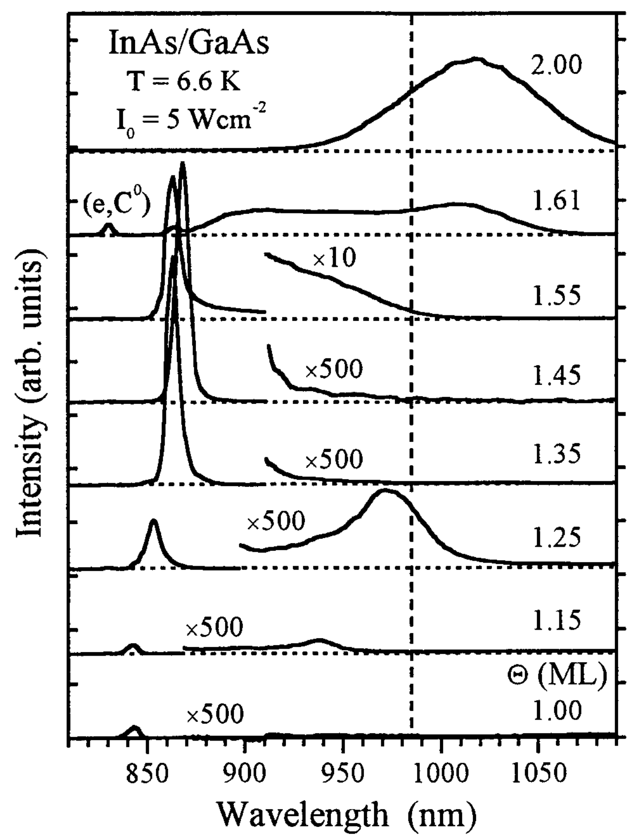

Fig. 3

insights into the long-standing problem of the kinetic aspects of 2D to 3D morphology change not embodied in the widely encountered Stranski-Krastanow growth mode. Moreover, this systematic study unambiguously identified the origin of the lasing in our InAs quantum box-based laser structures as arising from the 3D island quantum boxes and not from other structural features. (see point 5). The spectrum position of the observed lasing line (see point 5) is shown as a dashed straight line in Fig. 3 for later reference. Since the laser structures contain InAs layer(s) with 2ML InAs deposition for which the quasi 3D clusters have vanished, the origin of lasing is attributed to the well formed, coherent InAs islands.

3. Island induced adatom migration during cap layer growth:

   Through transmission electron microscope (TEM) studies of InAs island samples having GaAs cap layers containing AlGaAs marker layers we demonstrated InAs 3D island induced migration of Ga away from the islands during growth of the GaAs cap layers. A mechano-chemical surface chemical potential based theory for growth profile evolution was developed and used in conjunction with the TEM observations to estimate the spatial range of the island induced strain fields for the first time. This is reported in publication nos. 7, 9 and 11.

4. Demonstration of vertically self-organized growth:

   The above noted InAs island induced stress/strain fields in GaAs cap layers were exploited to demonstrate theoretically the kinetically controlled occurrence of vertically self-organized 3D island stacking in multi-layer growth and experimentally the realization of such vertically self-organized growth. These results are given in publication nos. 12 and 13. Figure 4 shows an illustrative cross-sectional TEM image of a stack of five InAs 3D island layers separated by 36 ML thick GaAs spacers and reveals the vertically self-organized growth.

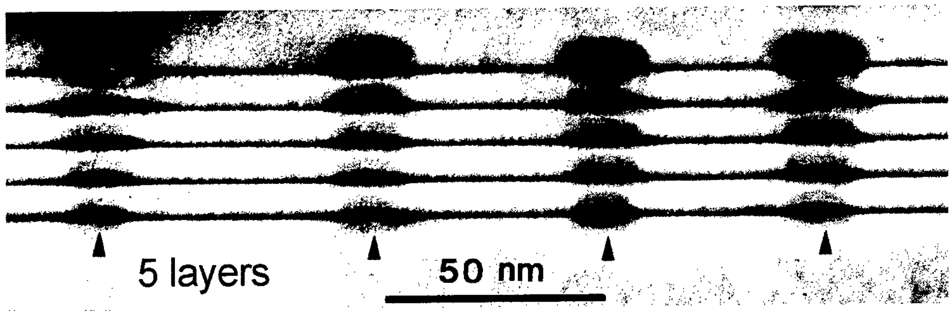

Fig. 4

5. Demonstration of Lasing from Vertically Self-Organized 3D Island Quantum Dots:

The results are shown in publication nos. 18 and 30. Ultra low threshold lasers are a critical component of high-density, high-throughput information processing systems. Owing to the discrete density of electronic states of an ideal quantum box, several over an order of magnitude type improvements in the figures of merit of devices based upon quantum boxes are theoretically expected. For lasers, these include threshold currents in the less than 10μA regime and high characteristic temperatures leading to much improved thermal stability.

Figure 5 shows a cross-sectional TEM image of a laser structure comprising five sets of vertically self-organized quantum dots as the active region sandwiched between $[(GaAs)_M/(AlAs)_N]_P$ based graded index optical confinement layers and AlGaAs cladding layers. Figure 6 shows the light output versus injected current behavior at 77K, indicating onset of lasing at a threshold current density (J_{th}) of ~310A/cm^2.

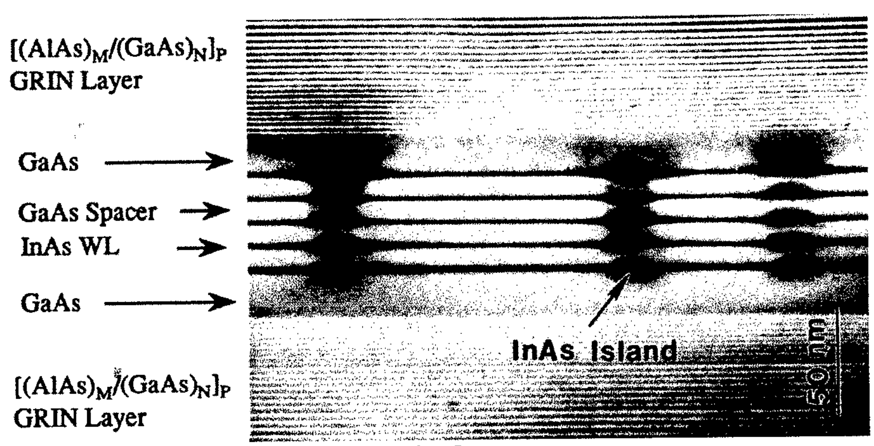

Fig. 5

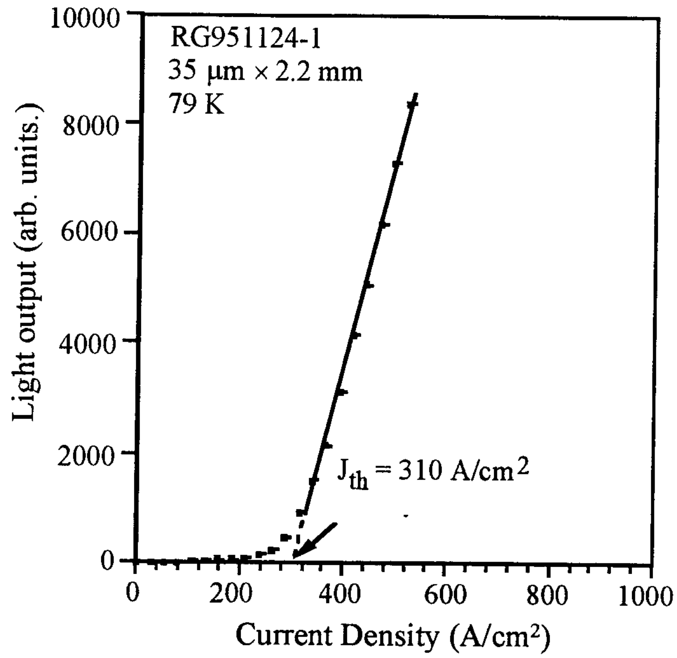

Fig. 6

II.2 Fabrication of Nanoscale GaAs Mesas and Growth of InAs on such Mesas

Control of the spatial distribution, the density and the size uniformity of the InAs islands is important to further exploitation of these island structures as quantum boxes for electronic and optoelectronic device applications. An important path towards these objectives is the use of patterned substrates containing mesas of appropriate sizes and profiles to achieve selective growth on the mesas. Proper exploitation of the lattice misfit strain for these objectives demands mesa widths in the sub-micron to sub 100nm regimes. We utilized our previously developed techniques of size-reducing epitaxy (publication nos. 1-3, 10, 14-15, 20, 22, 27-29) on *ex-situ* photolithographically patterned as well as *in-situ* direct write patterned stripe mesas of as-patterned width ~1µm to create the desired nanoscale mesas *in-situ* via purely growth control. A brief summary of the latter is given below as it is new and unique to our group. InAs island formation studies however, could be carried out only on the *ex-situ* as-patterned substrates due to both limitation of time and resources.

1. Growth of GaAs on *in-situ* patterned GaAs substrates:

As a first step towards an all UHV *in-situ* process for creating arrays of nanoscale mesas, we studied the growth of GaAs on patterned substrates prepared *in-situ* via focused ion beam (FIB) assisted Cl_2 etching of GaAs(001). FIB assisted Cl_2 etching technique is compatible with the UHV environment and hence is promising for an all *in-situ* approach to growth/processing/re-growth. The results are detailed in publication no. 26. The highly non-equilibrium nature of such FIB assisted gaseous dry etching also affords creating mesa sidewall orientations other than those provided by the thermodynamically most stable planes accompanying the usual wet chemical etching. An illustrative AFM image is shown in Fig. 7 for a stripe with ~26° sidewalls. Figure 8 shows the nature of the size-reducing growth on such mesa stripe as revealed by TEM images of growth of GaAs with AlGaAs (light bands) marker layers. We note that the as-patterned surface roughness of about 20nm is completely healed with typically about 50nm of GaAs buffer layer growth.

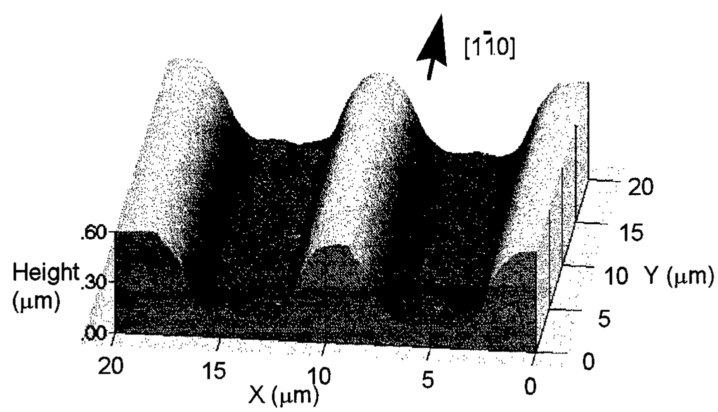

Fig. 7

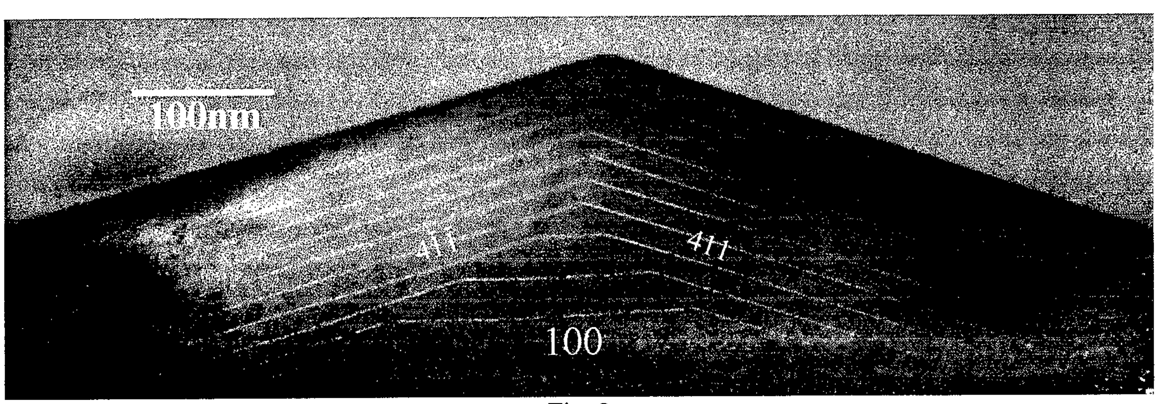

Fig. 8

2. Growth of InAs on GaAs nanoscale stripe mesas:

In Fig. 9 are shown some results of controlled deposition of InAs on nanoscale stripe mesas prepared *in-situ* via GaAs size-reducing epitaxy on *ex-situ* as-patterned mesas of widths ~1μm . Note that in these growths the InAs deposition amount is less than the critical deposition amount (1.57ML) for 3D island quantum dot formation on the planar substrate. The images show that, by controlling the strain and interfacet migration, it is possible to achieve selective area growth of InAs islands, in this case on the mesa top only, while also achieving control of the spatial distribution and density of the InAs islands. Note the reduction from three parallel rows of InAs 3D islands to one as the mesa width is reduced from ~100nm to 30nm. While some very promising results were obtained on this front, this contract expired and this approach could not be pursued much further. These results are a remarkable first and are to be published.

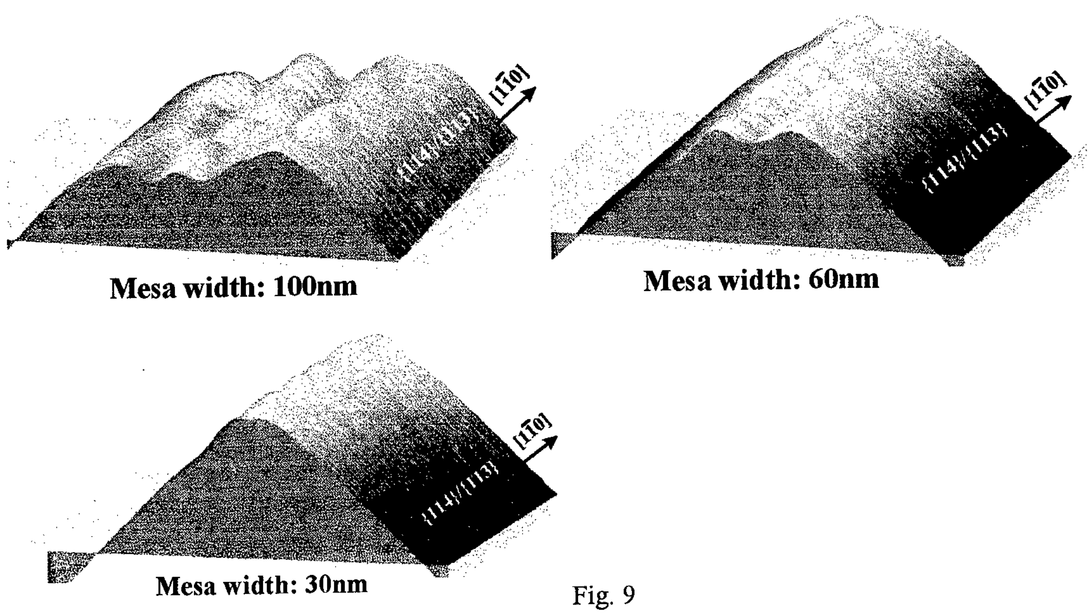

Fig. 9

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V. REPORT OF INVENTIONS: None